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INCORPORATING METEOROLOGICAL DATA INTO RECEPTOR ANALYSIS: RAPS REVISITED

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16. ABSTRACT

An analysis of data collected in the St. Louis Regional Air Pollution Study indicates that about 72% of the annual average PM-15 aerosol is background material, presumably of regional origin. The fine and coarse fractions are composed largely of sulfate and crustal material, respectively. About 29% of the particle mass is unaccounted for assuming the measured elements are present as stable oxides. The ratio between PM-15 and TSP concentrations varied considerably among sites depending on averaging time and percentile. Dispersion model results corroborate the regional origin of the bulk of the aerosol. Annual average dispersion modeling can reproduce the observed annual concentrations from local sources emitting trace elements; however only about 14% of the PM-15 mass can be accounted for by local point sources. A sensitivity analysis indicates that incorporating settling and deposition processes in dispersion models is unlikely to significantly alter predicted concentrations from local sources.

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INCORPORATING METEOROLOGICAL DATA INTO RECEPTOR ANALYSIS: -RAPS REVISITED

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An analysis of data collected in the St. Louis Regional Air Pollution Study indicates that about 72% of the annual average PM-15 aerosol is background material, presumably of regional origin. The fine and coarse fractions are composed largely of sulfate and crustal material, respectively. If it is assumed that the measured elements are present as stable oxides, then about 64% of the particle mass is accounted for. The ratio of PM-15 mass to TSP mass varied considerably among sites. Annual average pollutant roses for some elements show striking directional patterns which indicate the location and emissions of local sources. Sulfur and crustal elements exhibit more uniform patterns. The use of these patterns in conjunction with receptor models may increase the ability to resolve individual sources.

Dispersion model results corroborate the regional origin of the bulk of the aerosols. Annual average dispersion modeling can reproduce the observed concentrations and pollutant roses due to local point sources emitting trace elements; however only a small fraction of the PM-15 mass can be accounted for. A sensitivity analysis indicates that incorporating settling and deposition processes in dispersion models is unlikely to alter significantly predicted concentrations from local sources.

Introduction

This paper discusses some issues relating to the source apportionment of ambient respirable particle concentrations in urban areas. Many urban areas have a large number of both point and area sources, and high background levels of pollutants, such as sulfate (SO4), which are thought to be of regional origin. In such areas, source apportionment, that is, the quantitative determination of the contributing pollutant sources, may be difficult given limitations of available modeling techniques, model inputs and monitoring data. This paper describes an application of a new particle dispersion model and several statistical analyses using data collected for the Regional Air Pollution Study (RAPS) in St. Louis during 1974-7. A comparison is made with receptor model results, as are suggestions for utilizing meteorological information in conjunction with receptor models.

Several investigators have estimated source apportionment in the St. Louis area using a variety of receptor techniques, including chemical elemental balance regression (1); factor analysis (2-4); and target transformation factor analysis (5). These studies have determined that about 60% of the fine fraction mass (particles 0-2.5 um in diam) is SO4 related, from unspecified sources, and about 70% of the coarse fraction mass (2.5-15 um) is soil related, which can be classified by the predominant mineral type. Apportionment estimates were obtained for additional source classes, including crustal/fly ash, motor vehicle, smelter, refuse, steel and paint pigment.

The primary interest of this paper is to examine methods which can increase the resolution of source apportionments. Apportionments would be more useful if a larger number of source classes and individual sources (within classes) could be discerned. In addition, providing estimates of the local and background contribution of SO4 is of particular interest since it composes a large fraction of respirable particles.

Description of Data

At 10 of the RAPS sites, dichotomous (dichot) and high volume (hivol) samplers were deployed to collect particles in various size categories. (See Figure 1. Figure 1 does not include the northernmost and southernmost particle monitors.) Dichots were operated continuously and collected 6 or 12 hour samples, which were analyzed for total mass and 27 elements in fine and coarse fractions. The elemental analysis was determined by x-ray fluorescence, and the sensitivity and accuracy of the analysis varied depending on the element (6). The hivol samplers, which capture particles up to about 45 um in diameter, collected 24 hour samples every third day. Hivol filters were analyzed for total mass, SO4 and nitrate (NO3). Data from 1976 were used in this study because this year of RAPS had the most complete data capture.

The RAPS surface meteorological network consisted of 25 sites taking observations at heights of 10 and 30 m (Figure 1). These data were preprocessed into hourly observations of stability category, wind speed, wind direction and ambient temperature, using an average across the 25 sites. The upper air network in 1976 consisted of 3 sites where pibals and radiosondes were released 4 times daily. Upper air data were interpolated into hourly estimates of rural and urban mixing heights. The hourly data were further processed to obtain 12 hour averages (00:00 to 12:00, and 12:00

to 24:00) compatible with the dichot data. In addition, the circular dispersion of wind direction, a measure for directional data analogous to the standard deviation for linear data (7), was computed for each 12 hour period.

A comprehensive area and point source inventory was generated as part of RAPS. The point source inventory contained hourly estimates of emissions of 10 pollutants from over 300 point sources. Point sources were aggregated into 69 facilities, for which annual average emissions were determined. Facility parameters, i.e., stack height, cross-sectional area and flow rate, were determined by taking an emission weighted average of the stack parameters. The area source inventory contained hourly emission estimates from nearly 2000 sources.

Particle Composition

The annual average dichot composition averaged over the 10 monitors is presented in Table I. While 27 elements were measured on each filter, only the principal constituents are presented. The fine fraction is composed primarily of SO4; the coarse fraction consists mostly of crustal materials. Measured elements account for 21 and 44% of the fine and coarse mass, respectively. Assuming the elements are present in their oxidized form, 49 and 79% of the fine and coarse fractions, respectively, and 64% of the total PM-15 mass of 40 ug/m3, is accounted for. The remainder may be composed of carbonaceous material (graphitic carbon and hydrocarbons), water (hygroscopic and crystalline), and nitrogen compounds (NO3 and ammonia). The water content of the aerosol may be seasonal, depending on climatic factors. NO3 measured with hivols accounted for about 4.6% of TSP, and SO4/NO3 ratios were similar across monitors and averaged 3.4. Assuming the same average ratio in dichot samplers, NO3 would compose 6.2% of dichot mass. Because SO4 and NO3 measurements may have large artifacts (8,9), the NO3 apportionment is uncertain.

Background Estimates

The general procedures for estimating background concentrations use either the average of concentrations at monitors, "excluding values when the source in question is impacting the monitor," or monitoring at a "regional" site which is away from local sources, but impacted by similar sources (10). We have assumed that the background is the lowest concentration in the network at each time. The other procedures give similar background estimates. The annual average PM-15 background was found to be 29 ug/m3, representing about 72% of the total mass.

Continuous measurements of SO4 were not collected in RAPS, but comparisons between simultaneous hivol SO4 and dichot elemental S measurements showed high correlation, 0.53 to 0.94, depending on the monitoring site. Thus, elemental S is a good surrogate for SO4. The elemental S background averaged 2.2 ug/m3, about 76% of the annual average of 2.9 ug/m3. Assuming that all S is in the form of SO4, about one half of the average fine mass and about one quarter of PM-15 is composed of sulfate. However, hivol SO4 was an average of 34% higher than SO4 estimated using dichot S measurements, which is perhaps attributable to artifacts on the hivol filter substrate (9).

PM-15/TSP Relationship

Under the proposed PM-10 standards, more frequent monitoring is required in areas experiencing higher PM-10 concentrations (11). The proposed standards suggest estimating PM-10 using existing TSP data. Therefore, it is important to investigate the relationship between dichot and hivol measurements.

The fine and coarse fractions of the dichot samples were summed on days for which TSP measurements were available. A regression of TSP (as the dependent variable) and PM-15 was performed at each monitoring site (Table II). While the slopes at most sites are close to unity, the intercept terms are large, approximately half of the annual average TSP concentration. The R2 values indicate fairly low correlation, between 0.13 and 0.53, at the 10 sites. Urban sites had the highest correlation. Table III lists the ratios of PM-15 to TSP concentrations for annual and 24 hour averages, the latter expressed as the interquartile range (25th to 75th percentile) and the extreme range (1st to 99th percentile) of the ratios observed at the 10 sites. The ratios are based on dichot and hivol samples that are coincident in time. Annual average ratios ranged between 0.47 and 0.63. (Annual TSP levels ranged from 54 to 99 ug/m3 at the 10 sites). The ratios show greater variation for higher percentiles. For example, the average interquartile range at the 10 monitors was 0.42 to 0.68. (Highest daily TSP concentrations ranged from 152 to 483 ug/m3). While not necessarily representative of other locations, the data indicate a wide range of PM-15/TSP ratios.

Pace and Frank (12) proposed a methodology to estimate PM-10 levels from TSP data utilizing frequency distributions of the PM-10/TSP ratios. An analysis of 65 sites and 3 years of data from the EPA's National Inhalable Particulate Network has shown an annual average PM-15/TSP ratio of 0.55; an interquartile range of 0.48-0.62, and extreme range of 0.29-0.90. For 24 hour averages, the interquartile and extreme ranges were 0.46-0.63 and 0.21-0.92, respectively (13). The study also noted statistically different ratios between urban and rural sites and other factors. Using RAPS data, the same annual average ratio was found as in the nationwide study. The range for 24 hour averages was found to be comparable to that of the national study.

Dispersion Modeling

Long term pollutant concentrations were estimated using the new Particle Episodic Model (PEM) (14). PEM is a plume type model which incorporates deposition, gravitational settling and linear transformation processes. PEM calculates concentrations over the urban scale (less than 60 km) from area and point sources. With respect to deposition and settling, PEM provides greater flexibility and realism over comparable models.

The settling velocity w varies with particle density and diameter. For particles with diameters from about 2 to 50 um, w is about 1 cm/s, and about 1 to 3 orders of magnitude smaller for submicron size particles (15). Deposition velocity v is difficult to measure and the uncertainties are large. SO2 deposition velocity estimates vary from 0.04 to 7.5 cm/s (16). For large particles (greater than 50 um), gravitational settling is the primary removal mechanism and $v=w>0$. In the case of a gas with perfect reflection from surfaces, $v=w=0$.

A sensitivity analysis was performed to determine the effect of settling and deposition on ambient concentrations in the near field of a source (0-2 km). Figure 2 shows downwind concentrations from a point source with an effective height of 17 m and an emission rate of 1 g/s under 3 m/s winds in neutral (DD) stability. Deposition and settling velocities are varied over the range expected for PM-10. Concentrations near the source are seen to be insensitive over the expected range of v and w.

The effect of settling and deposition in middle to far field of a source (>2 km) was also investigated. For the source modeled above, a deposition velocity of 1 cm/s results in 74% of the concentration at 10 km, as compared to v=w=0, and only 33% at 60 km downwind. Thus, concentrations in the far field may be quite sensitive to deposition velocity. However, ambient concentrations will be very small at large downwind distances. In general, the effect of settling and deposition increases with larger w and v, lower wind speeds, more stable conditions and greater downwind distances.

The incorporation of deposition and settling processes in dispersion models is expected to produce small changes in PM-10 concentrations from local sources (i.e., within a few km), which are the primary contributors of ambient concentrations (excluding the background or regional component). Much longer distances are required for significant deposition and settling. Varying the settling and deposition velocities over their expected range altered aggregate concentrations by less than 1% at most receptors in urban applications of PEM.

The transformation of a local primary pollutant (SO2) to a secondary pollutant (SO4) may be modeled by PEM. While the ratio of SO4/SO2 increases with downwind distance from a point source, both concentrations decrease with distance due to dilution. Primary emissions of SO4 range from about 1 to 14% of SO2 emissions, depending on the source. For the entire St. Louis inventory, primary SO4 emissions have been estimated as 1.85% (17). Expected transformation rates are about 1% per hour. Thus, because several hours residence time is necessary for secondary SO4 from local sources to exceed primary SO4, little production of SO4 is expected given the short travel times of local pollutants to urban monitors under most meteorological conditions.

A procedure was developed to compute efficiently long term average concentrations using PEM. The contribution from each source was estimated as the sum of the products of 1) plume centerline source concentrations for each wind direction and stability class, 2) annual probabilities of the wind direction and stability combinations, and 3) smoothing factors particular to the stability category and the source-receptor distance for each direction-stability combination. The smoothing factor reflects the distribution of wind directions experienced during the 12 hour period of particle sampling. PEM was used to calculate hourly plume centerline concentrations in 7 stability categories at distances from 0.1 to 60 km, assuming average temperature, wind speed and mixing height in each stability category. The probability of all combinations of 16 wind sectors and 7 stability categories in St. Louis was determined for the 8774 hours in 1976. This procedure should not introduce significant biases with respect to wind speed and mixing height, since these variables tend to be isotropically distributed over the long periods modeled. The outputs of the long term procedure included 1) annual concentration levels, 2) source apportionment at each receptor; and 3) receptor concentrations stratified by wind sector and stability category.

Dispersion Model Application

In order to estimate the local and background contributions, dichot observations were compared to dispersion model predictions. Pollutant "roses" for observed levels of PM-15 at the 10 sites (located at their UTM coordinates) are shown in the first panel of Figure 3. Pollutant roses indicate concentration stratified by wind direction, where the length of the "arms" represents the average concentration for winds blowing from a given sector. The roses "point" to emission sources which produce higher pollutant concentrations at a sampling site. The second panel of Figure 3 shows observed PM-15 concentrations with the background subtracted. The background rose is shown in the fourth panel. (Note the differences in scale for each panel.)

TSP concentrations due to point sources only were predicted using the dispersion model and the long term procedure. Annual average predictions ranged from 1.5 to 10.5 ug/m3 at the 10 sites, and averaged 5.7 ug/m3. This is a small fraction of observed levels of PM-15 (29-59 ug/m3) and TSP (54-99 ug/m3).

Using estimates of PM-15 point source emissions, the third panel of Figure 3 displays the predicted PM-15 roses. If background is neglected, predicted and observed roses of PM-15 bear little resemblance. Accounting for background levels is very important in dispersion modeling.

Figure 4 shows pollutant roses for observed fine fraction elemental S and predicted SO2 roses, to which primary SO4 will be proportional. Again, these roses appear quite different. For SO2, PEM predictions of point source contributions range from an annual average of 30 to 90 ug/m3 over the receptor grid (average 40 ug/m3). Using the 10 site average and 1.85% primary sulfate estimate, annual average primary SO4 concentrations would be about 1.2 ug/m3. Even considering transformation, PEM predictions of SO4 from local sources will not approach the 11.7 ug/m3 observed at the hivol sites and the 8.7 ug/m3 average estimated from the dichot measurements.

Dispersion modeling can predict elemental concentrations and apportionment. This could provide an opportunity for model validation when there are few sources of some elements and low background levels. For example, the St. Louis airshed contains a single major source of Ti, a paint factory located just south of St. Louis. The background concentration of Ti is essentially zero. In Figure 5, the observed pollutant roses exhibit a striking directional pattern for Ti. Annual average Ti concentrations were calculated from the dispersion model using the long term procedure and a nominal emission rate of 1 mg/s. Figure 5 shows observed and predicted pollutant roses for fine and coarse Ti fractions. No deposition or settling was assumed for the fine fraction; settling and deposition velocities of 1 cm/s were used for the coarse fraction. Both predicted and observed roses clearly indicate the location of the Ti source.

Observed vs predicted annual fine fraction Ti concentrations showed good agreement among the 10 sites and a correlation coefficient of 0.718; coarse fraction variables have lower correlation (0.575). Incorporation of deposition and settling velocities in the coarse predictions marginally altered the correlation, although predicted concentrations decreased by about 30%. The emission rate of Ti was estimated by fitting a line to the scatterplot of observed vs predicted concentrations. Estimated fine and coarse fraction Ti emissions are 43 and 211 kg annually, respectively.

The Ti modeling results indicate the potential performance and utility of dispersion models for source apportionment. However, for many fine and coarse particles there are numerous emission sources and high background concentrations.

Dichot Concentrations Stratified by Wind Direction

Several methods were evaluated to determine the effect of wind direction during the 12 hour sampling periods of the dichot samplers. Pollutant averages stratified into 16 sectors were calculated using: 1) average resultant direction; 2) average direction using sampling periods with directional variability (dispersion) less than 10, 20, 40 degrees respectively; and 3) hourly direction data (assuming a constant pollutant flux in the 12 hour period). Restricting wind variability to 10 degrees eliminated 85% of the observations. With less stringent directional criteria, methods 1 and 2 gave comparable results. Method 3 tended to obscure directional effects. Despite these differences, there was little sensitivity to the choice of method in annual pollution roses, and concentrations were stratified using periods in which the directional variability was less than 40 degrees. This criterion eliminated approximately 15% of observations.

Figures 6a and b show roses for 20 fine fraction variables. Figure 7 shows coarse fraction variables. Roses were not calculated for elements that were not significantly above detection limits. Several fine and coarse fraction metals have very strong directional effects, including Ti, V, Ni, Cu, Cd, Sn and Zn. In contrast, Al, Si and S display isotropic distributions and similarly shaped roses at the 10 sites for each element, with the exception of the Si rose at site 3. This site also has a very sharp and unique rose for Mn. Coarse fraction S at site 1 also displays large directional effects. The pollutant roses also indicate the relative concentrations among the 10 monitoring sites: rural sites have very low concentrations of Ti, Cl, Cr, Mn, Cu, Br and Pb as compared to urban sites. Concentrations of P, V, Ni, Zn and Sn are also lower at rural sites, but less dramatically so. This pattern suggests that urban sources emit most of these pollutants. In contrast, Al, Si, S, K and Ca concentrations are nearly the same for rural and urban sites. Most of the S is due to long range transport; the other elements are found in crustal material.

Figure 1 shows the location of major industrial facilities in the greater St. Louis area. Some of the pollution roses clearly point to major emitters, especially if the emissions contain unique tracers. The following sources can be readily identified:

A paint factory located just south of the city is the major source of both fine and coarse Ti.

Two adjacent refineries to the NE of the city are the probable sources of V, especially noticeable at sites 6 and 9.

A primary Zn smelter is indicated just E of monitor 2.

An iron works about 1 km W of monitor 3 is the likely source of Mn.

Iron, steel and smelting industries are indicated by the highly directional roses for Ni, Cu, Cd and Sn, which point towards several clusters (SE and SW of St. Louis) of these industries. More specific source profiles information may help decompose these sources.

The vehicular traffic is reflected in the similar Br and Pb roses.

Winter salting on high volume high speed roads is indicated by the coarse Cl roses. Roses at sites 1, 3, 5, 6 and 8 are oriented to nearby interstate highways 40, 56 and 70.

Fine Cl roses at sites 1, 2 and 4, near the Mississippi River, indicate a possible source of Cl towards the river.

A fertilizer plant east of site 1 is indicated by the coarse P and K roses.

The S and fine mass pollutant roses are similar, which is expected since fine mass is composed primarily of SO4. Most SO4 is probably derived from both local and regional coal combustion. The largest SO2 source in St. Louis is a steel facility in Granite City, just east of the city; the largest SO2 point sources in the greater area are two coal power plants 60 km and 106 km SW of the city. Coal fly ash contains Si, Ca, Al, As, Se, and Pb, many of which are also emitted by other source classes. One of the better tracers for coal aerosols is Se. The Se roses appear to be composed of two parts: a low level isotropic component of approximate equal magnitude at all sites, and a higher level component in the directions of the steel and power plants. Roses for other fly ash constituents, i.e., fine Al, Si, and Ca, also show a SW orientation at all sites superimposed on an isotropic distribution, suggesting contributions from the power plants. However, the orientation of the fine mass and S is from the east, in the direction of the Ohio River Valley area.

Conclusion

The examination of St. Louis aerosol data indicates that on an annual basis, about one half of the fine fraction particles and about one quarter of PM-15 consists of SO4. Receptor model studies (1-4) estimated 60% of the fine fraction was attributable to SO4, but these studies used only summer data when SO4 conversion is higher. Dispersion modeling of local sources, background estimates and examination of pollutant roses all indicate a regional origin of most of the SO4. Dispersion modeling of local point sources accounts for only a small fraction of the annual PM-15 concentration in the urban area. Annual average dispersion modeling may yield useful predictions of elemental apportionment, and can provide a way to estimate source emissions of unique aerosol constituents. The location of local sources of pollutants such as Ti, V, Ni, Cu, Cd, Sn and Zn can be identified using pollutant roses derived from 12 hour aerosol samples. In contrast, Al, Si, Ca and S are nearly isotropically distributed, indicating a wide distribution of sources or a long distance (regional) origin. The azimuthal distribution of elements might be combined with a knowledge of source compositions to better estimate apportionment and identify specific sources.

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Table I. Annual average mass and fractionization at 10 RAPS sites (in ng/m3, except total mass in ug/m3). Based on approximately 732 12 hour observations at each of 10 sites.

-elemental mass-			----------oxidized form----------				
element	fine	coarse	compound	fine	percent	coarse	percent
Al	119.4	914.1	Al2O3	225.5	1.11	1726.5	8.60
Si	242.2	3355.6	SiO2	519.1	2.57	7190.6	35.83
P	18.8	209.2	PO4	57.6	0.28	641.1	3.19
S	2602.6	284.2	SO4	7807.9	38.60	852.5	4.25
Cl	88.0	245.2	NaCl	145.1	0.72	404.0	2.01
K	150.6	313.3	K	150.6	0.74	313.3	1.56
Ca	124.9	2269.9	CaO	174.9	0.86	3177.0	15.06
Ti	28.0	147.6	TiO2	46.8	0.23	246.5	1.23
Fe	172.7	873.0	Fe2O3	246.6	1.22	1246.6	6.21
Pb	461.3	125.1	Pb	461.3	2.46	125.1	0.67
others	269.3	152.2	others	269.3	1.33	152.2	0.76
unknown	16191.4	11362.9	unknown	10364.6	51.21	4176.7	21.38
total mass	20.2	20.1	total mass	20.2	100.00	20.1	100.00

Table II. Regression of TSP on PM-15. Approximately 120 observations were available at each site. All coefficients are sig-nificant at the 99% level except the intercept for site 5 (97% level).

	regression coefficients		
site number	slope	intercept	R2
1	0.90	49.4	0.33
2	0.94	50.3	0.43
3	0.96	32.8	0.44
4	1.15	38.9	0.26
5	1.52	14.9	0.53
6	1.01	21.3	0.53
7	0.96	35.8	0.28
8	0.61	34.1	0.21
9	0.86	33.1	0.13
10	0.86	28.1	0.22

Table III. Ratios of PM-15 to TSP mass for 1976 at the 10 RAPS sites.

site number	ratio of annual average	interquartile range for 24 hour averages	extreme range for 24 hour averages
1	0.55	0.41 - 0.66	0.21 - 0.92
2	0.47	0.32 - 0.56	0.20 - 0.99
3	0.61	0.50 - 0.74	0.26 - 0.90
4	0.50	0.42 - 0.65	0.12 - 1.00
5	0.53	0.43 - 0.62	0.18 - 0.99
6	0.63	0.49 - 0.71	0.22 - 0.99
7	0.50	0.37 - 0.61	0.11 - 0.92
8	0.61	0.44 - 0.76	0.13 - 1.00
9	0.52	0.45 - 0.66	0.03 - 1.00
10	0.56	0.40 - 0.78	0.17 - 1.00
average	0.55	0.42 - 0.68	0.17 - 0.97

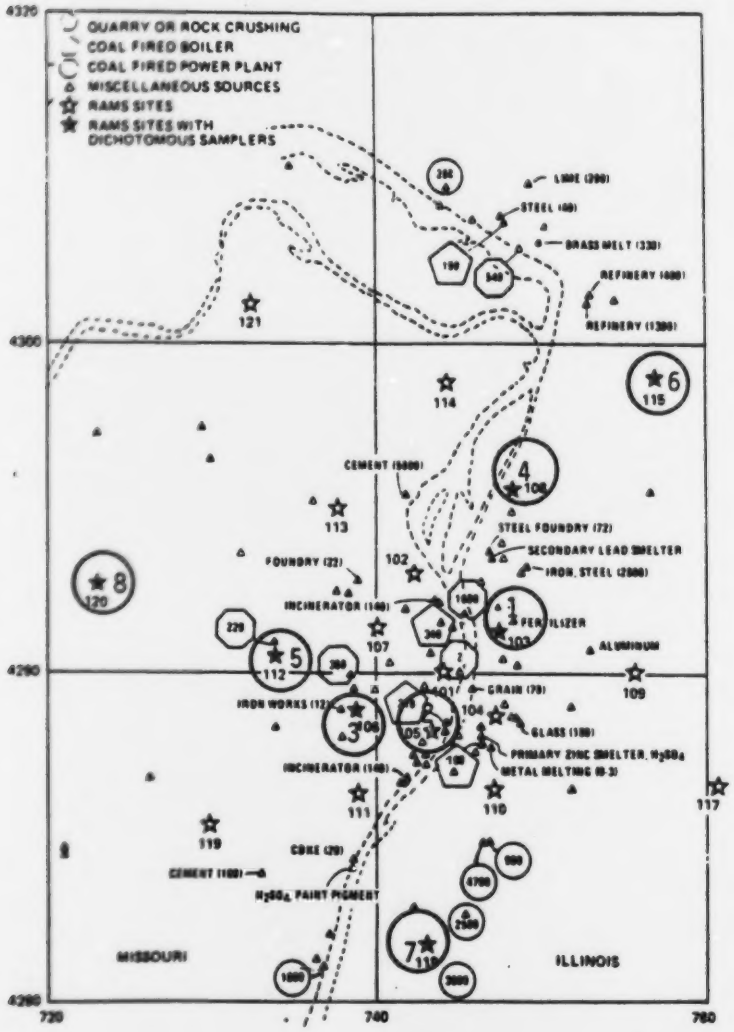

Figure 1. Location of monitors and major emission sources in the St. Louis area. Numbers in large circles indicate monitor sites. Emission rates are shown in tons per year in parentheses or within the symbols for selected sources. Sites 9 and 10 lie outside this figure at UTM coordinates (741.6, 4329.2) and (749.3, 4236.5). (1 UTM unit=1km.) Taken from Dzubay (1).

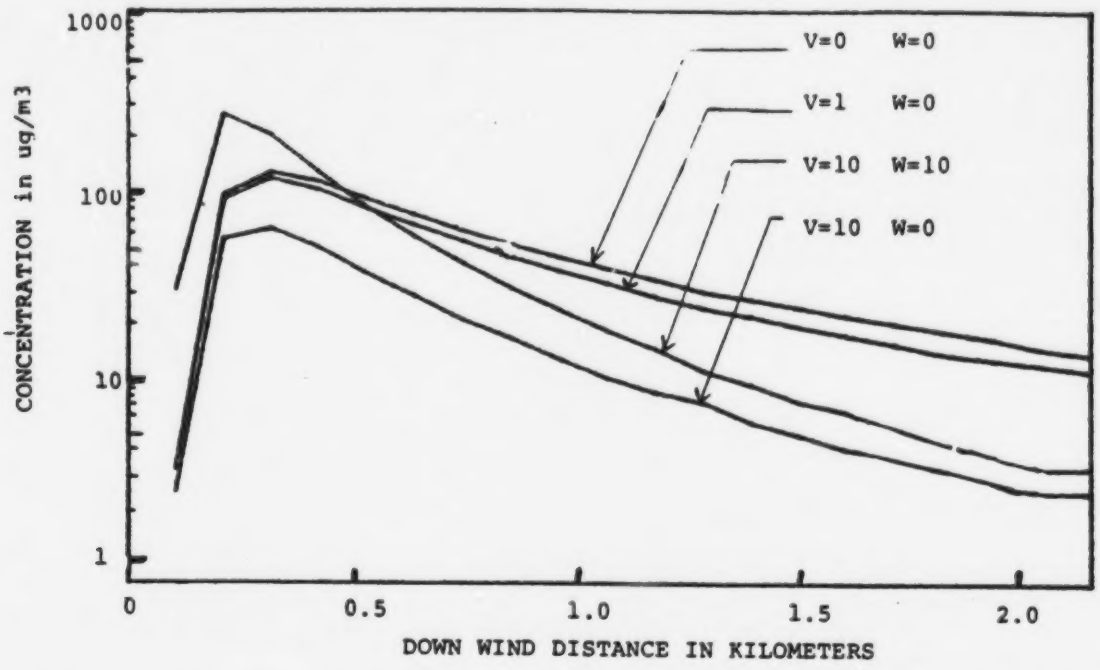

Figure 2. Ambient downwind concentrations from test source with specified deposition and settling velocities.

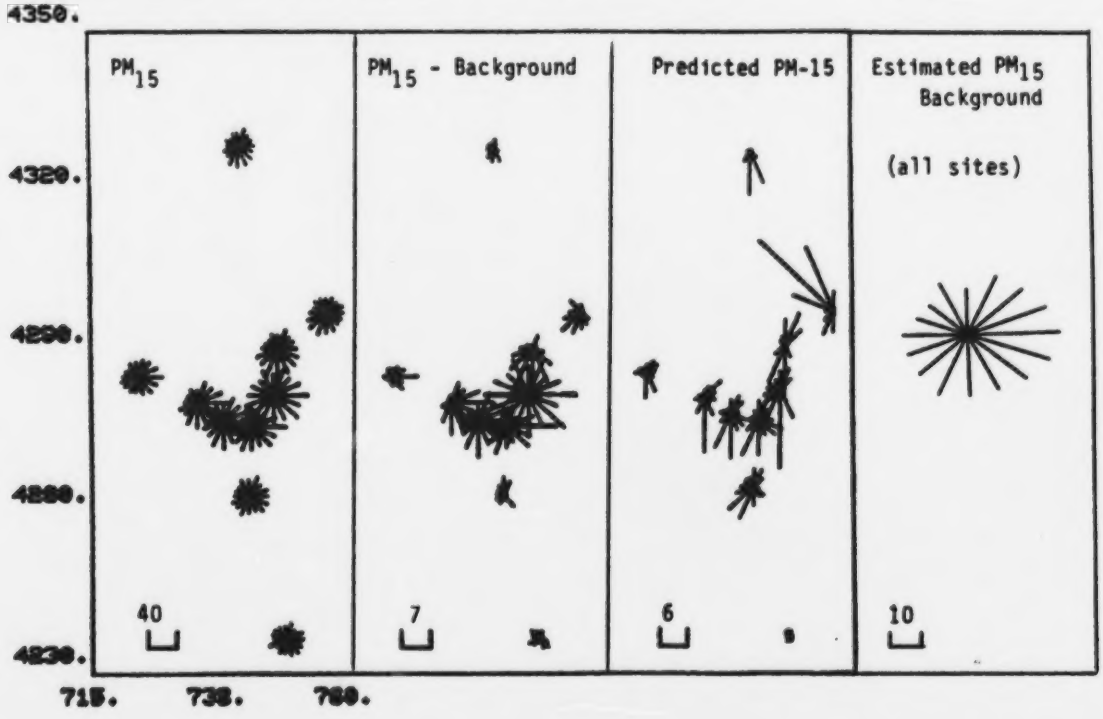

Figure 3. Observed and predicted pollutant roses of PM-15 at 10 particle monitors. UTM (Universal Transverse Coordinates) on axes. Roses indicate concentrations in 16 wind sectors. Scale in ug/m3.

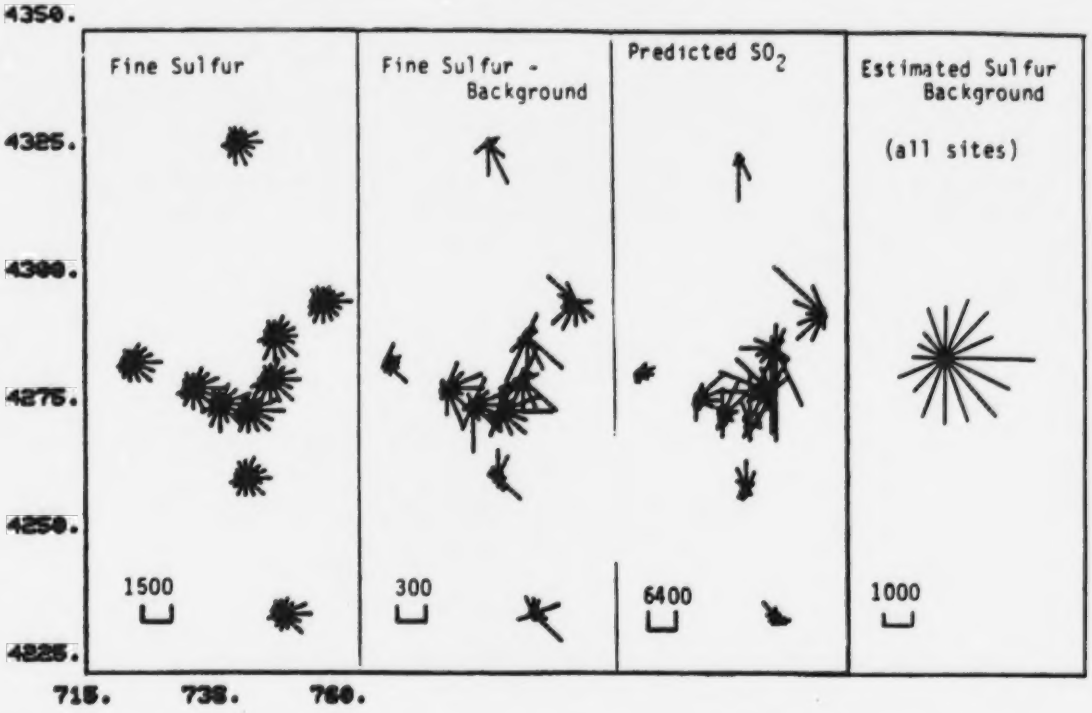

Figure 4. Observed and predicted pollutant roses of S and SO2. Scales in ng/m3.

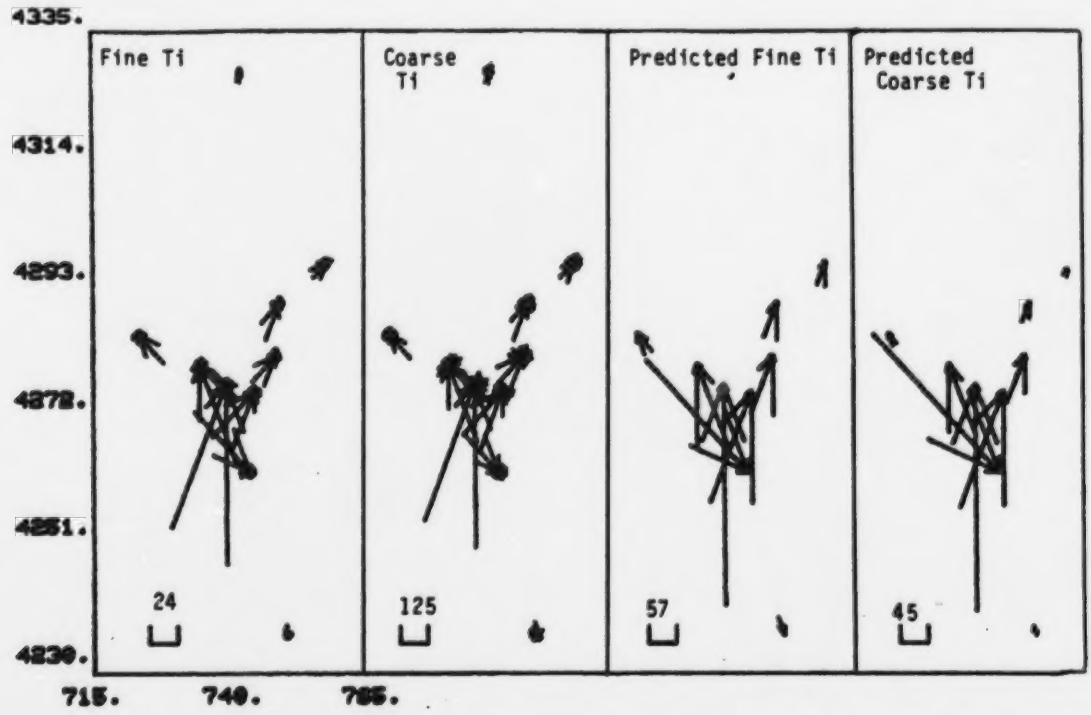

Figure 5. Observed and predicted pollutant roses of Ti. Scales in ng/m3.

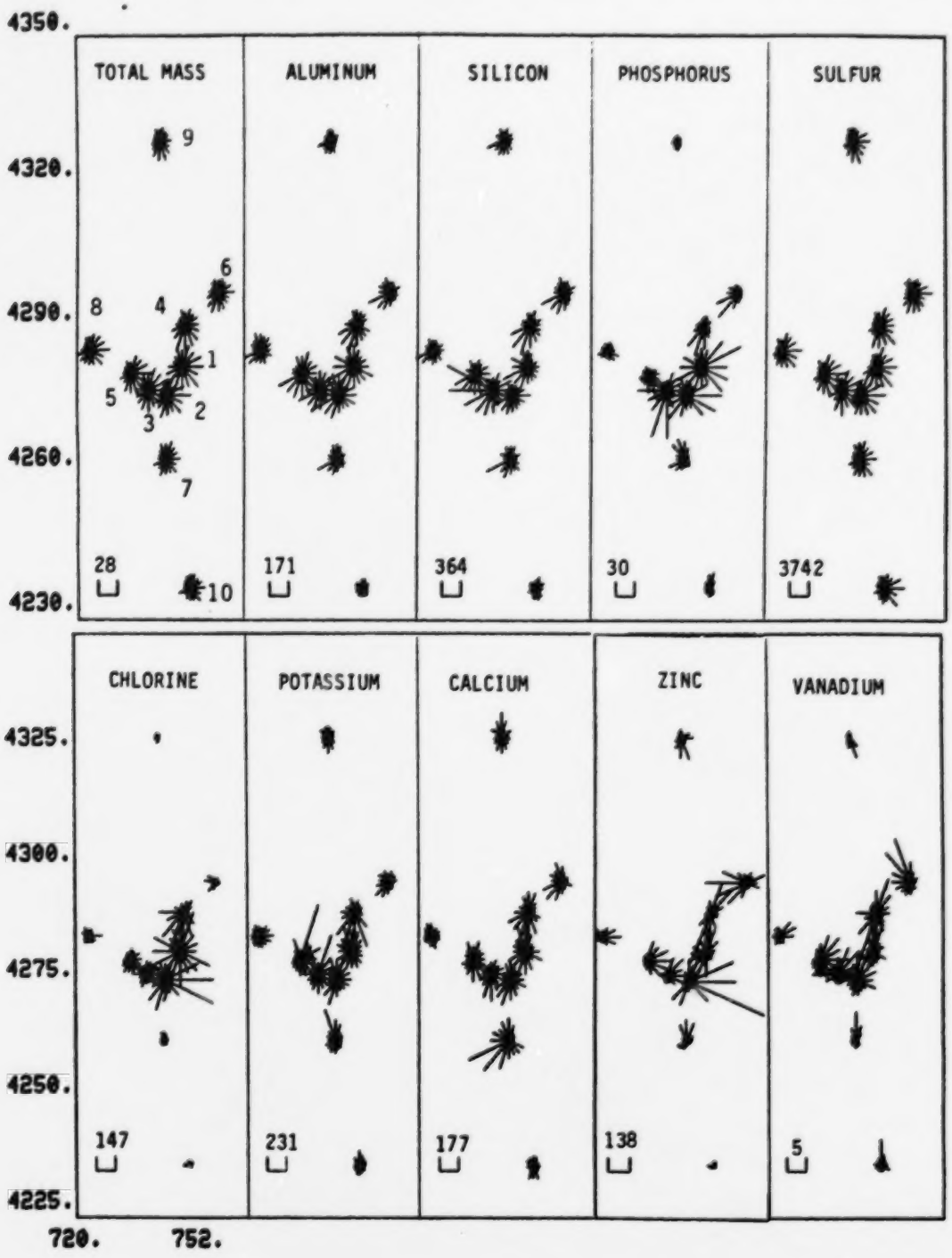

Figure 6a. Annual pollution roses for dichot fine fraction variables. Numbers indicate site number. Scales indicate concentrations in ng/m3, except total mass, in ug/m3. UTM coordinates on axes.

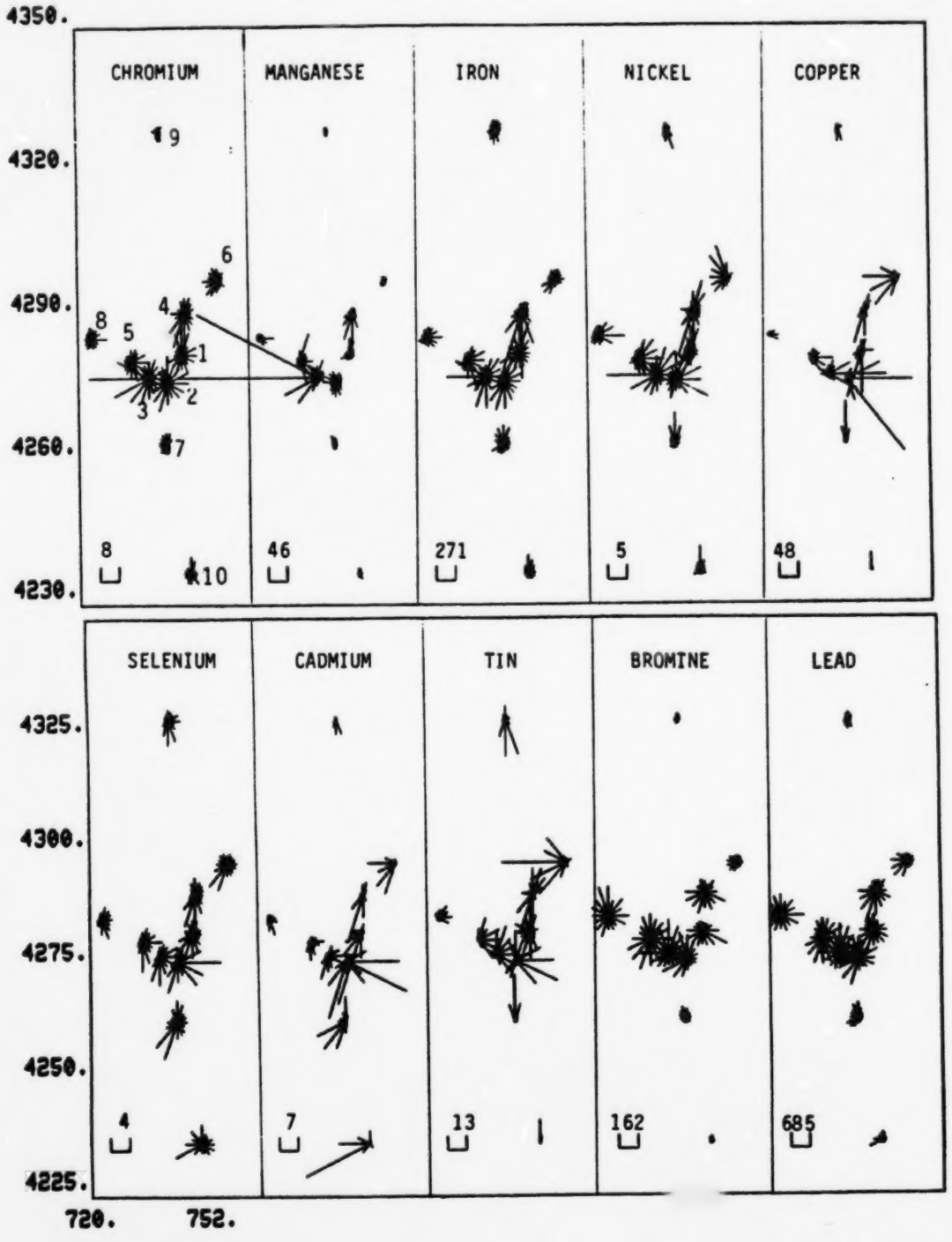

Figure 6b. Annual pollution roses (continued).

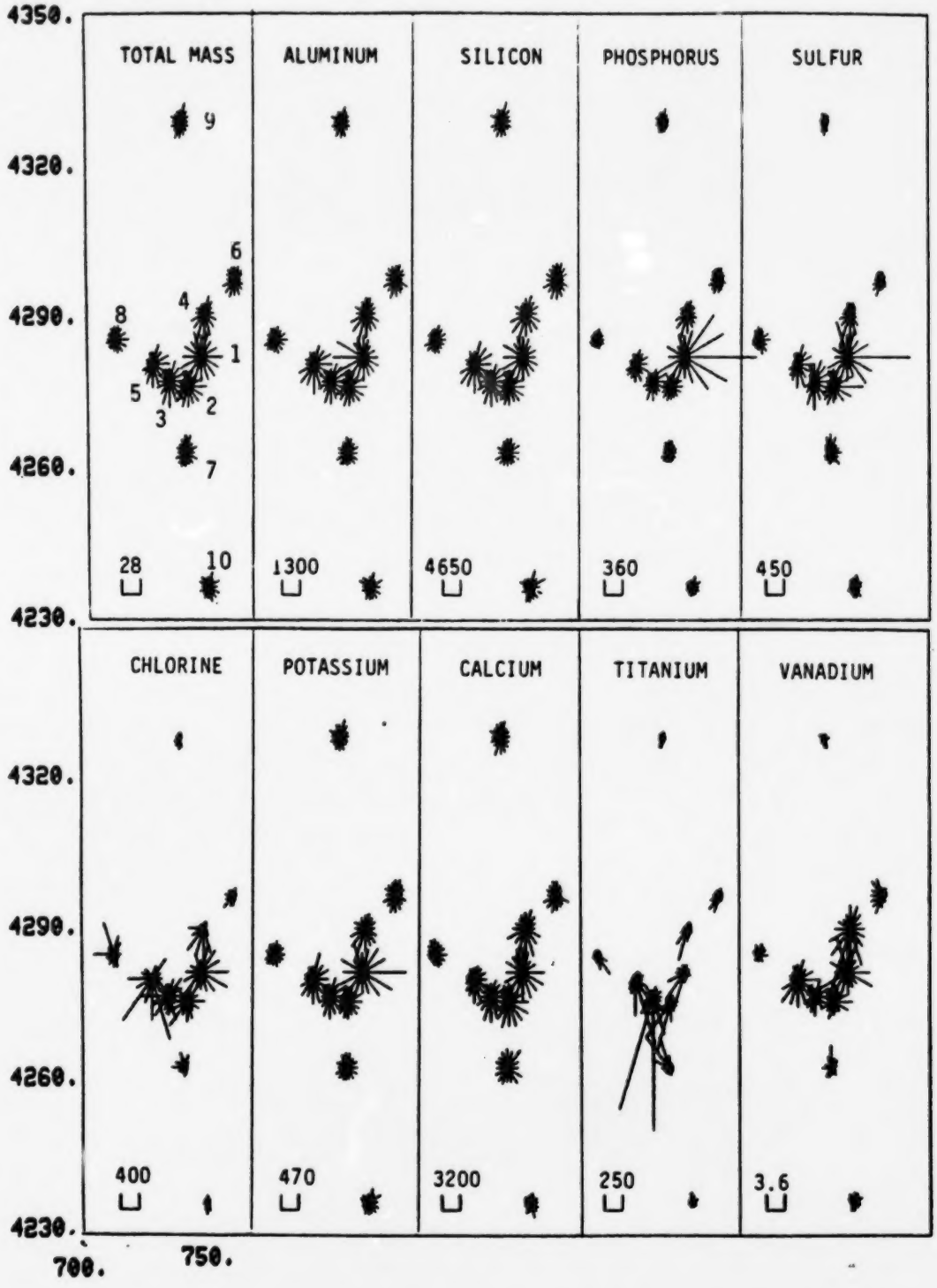

Figure 7. Annual pollution roses for dichot coarse fraction variables. Otherwise as Figure 6.